Intracavity absorption spectroscopy with a turbulent detuned actively mode-locked Ti:sapphire laser

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Abstract: Intracavity laser absorption spectroscopy (ICLAS) is an extremely sensitive method for the detection of very weak absorptions. However, the conventional use of multimode lasers has thus far significantly reduced its ability to detect in situ molecules and its sensitivity. We propose the use of a new type of laser that overcomes these limitations: the turbulent detuned actively mode-locked (TDAM) Ti:sapphire laser, which owing to its short coherence length, eliminates harmful intracavity interferences. The proposed technique called TDAM-ICLAS is furthermore highly sensitive to intracavity absorption, continuously tunable and has no frequency chirp.

OCIS codes: (010.1120) Air pollution monitoring; (300.1030) Absorption; (300.6360) Spectroscopy, laser; (140.3590) Lasers, titanium; (140.4050) Mode-locked lasers; (140.3518) Lasers, frequency modulated.

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1. Introduction

In situ and real-time measurement of small quantities of air pollutants is a major challenge for the monitoring and understanding of atmospheric chemistry. For in situ measurements the ICLAS (intracavity laser absorption spectroscopy) technique [1,2] has been superseded by the CRDS technique (cavity ring down spectroscopy). For measurements in gas cells, CRDS and its derivatives are now reaching sensitivities of $10^{-10}/\text{cm}/\sqrt{Hz}$ [3,4], while the sensibility of ICLAS capped at $10^{-9}/\text{cm}/\sqrt{Hz}$ [5]. Yet ICLAS is simpler and has significant advantages: the alignment of a single relatively low-finesse cavity, it is broadband, spectrally multiplex, and fast. A single Ti:sapphire laser can record spectra in the spectral range of 700-950 nm in time scales consistent with atmospheric fluctuations. In practice, ICLAS has been used with high sensitivity in gas cells, but only rarely in situ.

The use of multimode lasers induces intrinsic spectral noise (parasitic Fabry-Perot, birefringence of the amplifying medium, etc.) to the ICLAS [1,6,7]. The relatively long coherence length of multimode lasers cause a sensitivity several orders of magnitude lower than the theoretical limit of ICLAS [1,6].

In this work, we propose a solution that solves many of the problems of ICLAS and thus improves on the current limits. The next section describes the experimental setup. The following three parts compare and discuss ICLAS for three types of laser operation: multimode, frequency shifted feedback (FSF), and turbulent detuned actively mode-locked (TDAM). To our knowledge, the last two types of lasers are considered for the first time for ICLAS with solid state Ti:sapphire laser.

2. Experiment

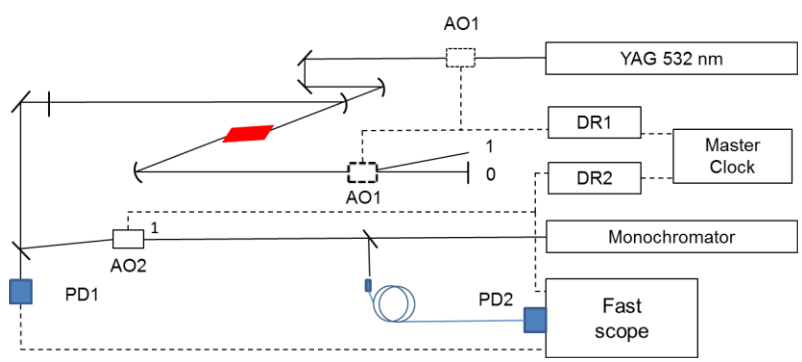

Fig. 1. Experimental set-up. See text.

The experimental setup is depicted in Fig. 1. We used a modified version of the Spectra-Physics 3900S Ti:sapphire laser pumped by a frequency doubled YAG laser (VERDI 5W). The high refractive index (2.25 at 800 nm) of the TeO_2 acousto-optic modulator (AO1) and the properties of the acoustic index grating required an accurate calculation of the cavity to operate the laser at the maximum of stability and to simplify adjustments. The angle and

length of the Z arm of the cavity containing the AO1 have been significantly modified. The cavity is coupled, according to the needs of the experiment, on either the first or zeroth order of diffraction of the AO1. On the first order, the laser yields a pumping threshold of 1.5 W, and all experiments were conducted with a pump power of about 3 W.
The AO1 modulator operates at a RF frequency ν_{AO} of 35 MHz, controlled via the driver DR1 by a master clock (Thales, tuning accuracy of 1 ns and typical jitter of less than 50 ps). The AO1 modulator can also be used in the YAG pump beam, for multimode operation. For frequency shifted feedback or turbulent detuned actively mode-locked operation, the AO1 is placed intracavity on either the first or zeroth order of diffraction, respectively.

The acousto-optic modulator AO2 opens a time gate whose width δt_g can be as low as 100 ps and whose generation time t_g is continuously adjustable. The modulators AO1 and AO2 are synchronized with the master clock. The refracted beam on the first order of AO2 is spectrally and temporally analyzed by a high-resolution monochromator (ultimate resolution ~0.1 cm^{-1}) and a fast fibered photo diode PD2 (Thorlab, 100 ps). The photodiode PD1 (Thorlab, 1 ns) monitors the temporal evolution of the laser intensity.

3. ICLAS with a multimode Ti:sapphire laser

The ICLAS technique has so far used the remarkable properties of multimode cw lasers with a high number of modes. When absorption is localized on a group of modes, or even on a single mode, the intracavity energy is redistributed over all other modes. The corresponding mechanism is complex and reviewed in [1]. The performance of an ideal Ti:sapphire intracavity spectrometer which has been evaluated [6] gives a maximum effective path length of 200 000 km and a sensitivity of 10^{-12}/cm/$\sqrt{\mathrm{Hz}}$. Whatever the type of multimode laser used, this limit has never been reached and the results in the literature gives the best experimental sensitivities around 10^{-9}/cm/$\sqrt{\mathrm{Hz}}$. One of the main reasons behind this discrepancy is related to the long coherence length of the laser modes and to birefringence effects. These effects are even more important with a solid gain medium such as Ti:sapphire.

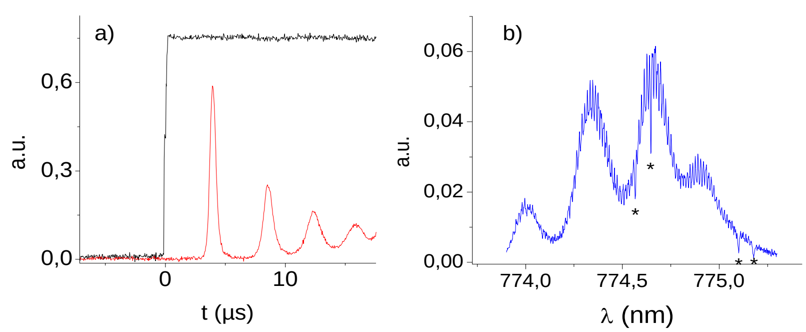

Fig. 2. a) Temporal evolution of the laser intensity (red) and AO1-RF command (black). The delay between rising edges is due to electronics and built-up time. b) Atmospheric ICLAS spectrum of the multimode Ti:sapphire laser around 744.6 nm. The stars mark O_2 lines.

For multimode operation, the AO1 is placed in the path of the pump beam and gated. The AO2 cuts a time gate that is analyzed by the monochromator. We recall that our cavity was not optimized for multimode operation (it has standard mirrors and optical axis, a linear cavity, etc.). Figure 2 shows the time evolution (a) and spectrum (b) centered at 774.6 nm for a generation time of 20 µs and an integration time δt_g of 2 µs. Microsecond temporal oscillations observed after the build-up are well known. Fig. 2(b) shows a very disturbed spectral envelope that is extremely sensitive to the adjustment of the cavity [6,7]. The rapid modulation (~8 GHz) corresponds to interference between the two faces of the output mirror. We used standard mirrors. In the literature, authors typically use special mirrors having an outer face cut to few degrees (up to the Brewster angle) with anti-reflective coating. This greatly limits the modulation effect observed here but does not eliminate it. The wider modulation (~150 GHz) is an interference effect due to the birefringence of the Ti:sapphire crystal. A very small misalignment with the optical axis of the crystal is almost inevitable. Hence, different crystal geometry where the optical C axis is parallel to the beam axis has been proposed [1,6]. The misalignment induces a lower birefringence, but the gain is half, so a higher pump power is needed which leads to other problems. Figure 2(b) also shows some absorption lines of O_2 embedded in the interference patterns.

Despite extensive precautions to mitigate these parasites, recent studies show a spectral envelope that still is very chaotic. For example, Fig. 2 of [7] shows the spectrum of a specially designed Ti:sapphire ring laser and Fig. 3(a) of [8] shows the spectrum of an Er^{3+} doped fiber laser. It is therefore necessary to correct the baseline. To do this one must eliminate the molecule in the cavity, record the baseline and then make the correction hoping that the adjustment of the cavity does not change between the manipulations [5]. This was achieved routinely in gas cells because operations can be done in a relatively short time. In general, it is not possible to remove a molecule quickly and easily for atmospheric in situ measurements.

4. ICLAS with a FSF Ti:sapphire laser

An idea for avoiding intracavity interference noise would be to introduce the modulator AO1 in the cavity of the Ti:sapphire laser and to close the cavity on the first order of diffraction. The laser thus formed is called a frequency shifted feedback (FSF) or modeless laser. This type of laser has remarkable properties that have been proposed for various applications, e.g., telemetry over long distances [9,10] and optimization of a laser guide star [11]. The emission characteristics of the FSF laser differ significantly from those of conventional lasers. A FSF laser intracavity optical wave ν_0 is shifted to $\nu_0+2\nu_{AO}$ after a round trip through the AO1 shifter, where ν_{AO} denotes the frequency of the corresponding acoustic wave. This continuous change in frequency disrupts the constructive interference of the fixed modes in a conventional laser. The transducer of the AO1 imposes a phase, which induces a spectral coherence over the whole laser spectrum [10]. From a spectral point of view, the FSF laser behaves as if all the frequencies contained in the optical spectral width were available. It is tempting to use a FSF laser to eliminate the harmful interference observed in the case of multimode lasers.

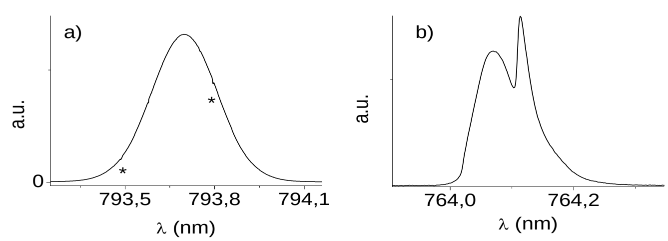

Fig. 3. a) Spectrum of the FSF-Ti:sapphire laser in a free absorption spectral range (t_g ~ 5 µs). Stars mark the imperfections of the photodiode array. b) Absorption spectrum of the O_2 molecule (t_g ~ 10 µs).

This is indeed what is observed in the spectrum of Fig. 3(a); the baseline is remarkably smooth. Only the AO1 was introduced into the cavity, no further optical element has been modified. The round-trip frequency shift $2\nu_{AO}$ not only prevents the constructive interferences which lead to the cavity modes, as has been demonstrated in the literature, but it also prevents the harmful interference induced by the faces of the mirrors and the birefringence of the crystal . The shape of the baseline is close to a Gaussian and is very reproducible. It can be easily corrected without removing the molecule to be studied.

Unfortunately, as can be seen in Fig. 3(b), the absorption line of O_2 is strongly deformed and enlarged. This is due to the frequency chirp (~15 MHz/ns). Photons undergoing absorption at a time t are frequency shifted with each round trip in the cavity. The absorption line then spreads to blue if the cavity is closed on the first order of the AO1, leaving a trail. A numerical calculation was conducted using the model in [12] with parameters adapted to the case of the Ti:sapphire laser. The losses are written:

$$\gamma(\nu) = \frac{1}{\tau_{RT}}\left(T + 2\frac{\pi\nu}{\Delta\nu_{AO}} + D\right) + \gamma_{abs}(\nu), \tag{1}$$

where, T is the output mirror transmission, $\Delta\nu_{AO}$ the spectral bandwidths of the AO1, D additional losses, and γ_{abs} the molecular absorption that can be written:

$$\gamma_{abs}(\nu) = \frac{2}{\tau_{RT}}\log(1 + A_{abs}\exp(-4\log 2(\nu/\delta\nu_{abs})^2)), \tag{2}$$

where, A_{abs} is the absorption in a round trip and $\delta\nu_{abs}$ the width (FWHM) of the molecular absorption line (typically 1.9 GHz for oxygen in normal atmospheric conditions). Calculation of the spectral broadening versus time, for the line of the oxygen molecule observed in Fig. 3(b) gives a width which increases with a rate of about 1 GHz/µs. This value is consistent with the experimental linewidth of about 9.6 GHz (FWHM) observed in Fig. 3(b) after 10 µs.

In an experiment with a dye laser whose output beam is slightly extracavity frequency shifted (<30 kHz) and fed back into the cavity, it has been shown that the sensitivity of ICLAS is already significantly altered [13]. In our case the intracavity frequency shift is 70 MHz (at each cavity round trip) that is much larger than the width of the free running laser modes. The dynamics is then very different, but the result is that the enormous frequency chirp makes ICLAS with a FSF laser unusable for dense spectra and long generation times. We could not detect the H_2O lines observed in the next section.

5. ICLAS with a TDAM Ti:sapphire laser

In general, an actively mode-locked laser is obtained in the configuration of the preceding paragraph when $2\nu_{AO}$ is equal to, with high precision, the free spectral range ν_{RT} of the cavity or possibly to a submultiple. The frequency tuning is generally maintained dynamically via a feedback. Otherwise, it is called a detuned actively mode-locked laser. As in hydrodynamics, it has been shown [14] that a first order transition from a steady state to a turbulent state appears when the detuning exceeds a critical value $\Delta\nu_c$. In the case of our Ti:sapphire laser $\Delta\nu_c$ is below 1 MHz.

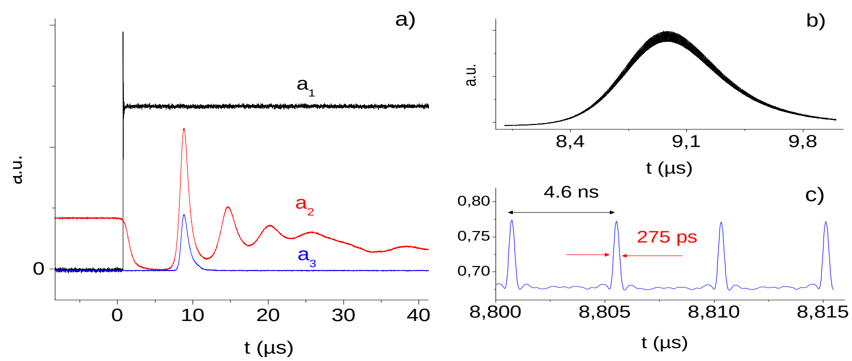

Fig. 4. Temporal evolution of the turbulent detuned actively mode-locked (TDAM) laser. a_1: AO1-RF command, a_2: output laser intensity from PD1, a_3: AO2 gated laser intensity. b) slow and c) fast time scale, showing comb and individual picosecond pulses.

The actively mode-locked situation is not observed in the configuration of our FSF laser when the cavity is closed on the first diffraction order of the AO1 because some parameters, e.g., losses and detuning are not adapted. We then switched to a new configuration: coupling the cavity on the zeroth order of diffraction of the AO1, gated as in the preceding paragraphs. When the radio frequency ν_{AO} is on, the dynamics of the laser is surprising. Figures 4(a)-4(c) show the evolution of the laser intensity on the three time scales observed using PD1 (ns) and PD2 (100 ps). On the rising edge of the acousto-optic modulator command (curve a_1), when the traveling acoustic wave starts, the strong perturbation generated turns off the laser (curve a_2). There is then a built-up followed by microseconds oscillations similar to the two previous cases. On a picosecond time scale (Fig. 4(c)) there is a comb of picoseconds pulses (275 ps wide, limited by the response of the measuring system) with a repetition period of $1/\nu_{RT}$ (4.6 ns). These pulses are built on an apparently continuous background (Fig. 4(b)). This is the typical situation of turbulent detuned actively mode-locked lasers. ν_{RT} is about 219 MHz and $3\times(2\nu_{AO})$ is 210 MHz. This corresponds to a detuning of the cavity length of about 30 mm. We are therefore in the turbulent regime. When adjusting the length of the cavity (which was checked), in the absence of feedback of the acoustic frequency, the system remains mostly in the turbulent regime because the spectral jitter is a few MHz. The study of the corresponding dynamics will be reported in another article.

We focus this article on spectral aspect and detection of molecules in situ. Figure 5(a) shows a spectrum of the oxygen molecule around 767.5 nm as a function of the generation time t_g. The remarkable result is that the baseline is perfectly smooth, reproducible, and close to a Gaussian shape. Since the TDAM laser coherence length is of the order of millimeters, there is no build-up of harmful interference. The near-Gaussian baseline can be deconvoluted for in situ experiments. Another remarkable property is that the ICLAS measurement can be made for generation time t_g much lower than those used by other authors in the case of a multimode laser, which is rarely less than 100 microseconds. Figure 5(b) shows a linear evolution of an H_2O ICLAS signal for t_g = 5–55 µs (t_0 ~ 5 µs). Note that the time scale of the microsecond oscillations (Fig. 4(a_2)) is perfectly usable.

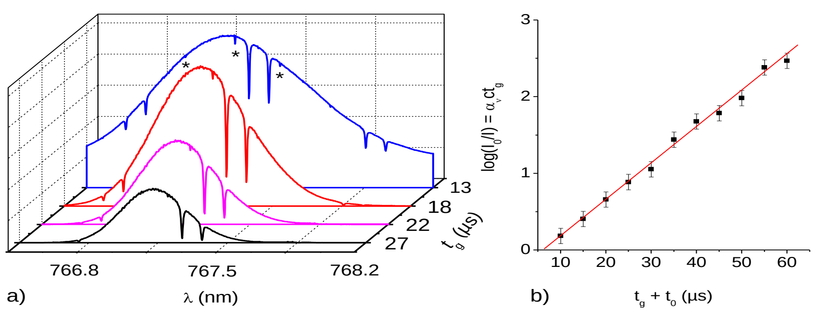

Fig. 5. a) Spectrum of atmospheric O_2 as a function of generation time. The stars mark imperfections of the photodiode array. b) Variation of the logarithmic ratio of the intensity of the baseline (I_0) by the intensity centered on an absorption line of the H_2O molecule (I) at the wavelength of 792.3 nm. t_0 is a delay due to the electronics and build-up and α_ν the H_2O absorption coefficient. The equivalent optical path length ($c.t_g$) varies from 1 to 20 km.

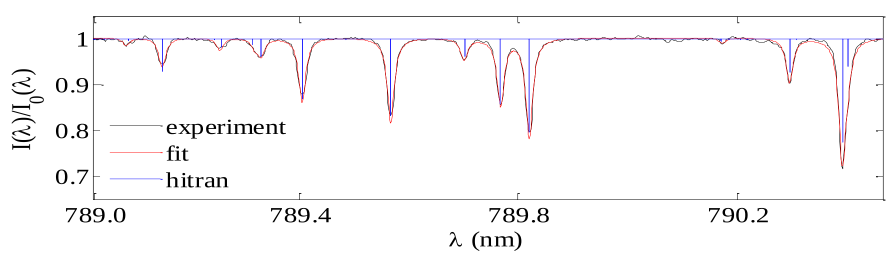

Fig. 6. In situ H_2O turbulent detuned actively mode-locked laser spectrum (TDAM-ICLAS).

Figure 6 shows an absorption spectrum of the H_2O molecule in the atmosphere of the laboratory for a generation time t_g of 10 µs, a AO2 time gate δt_g of 1 µs and an integration time of the monochromator detector of about 100 ms. The baseline spectrum was deconvoluted using a homemade fit (Matlab) which uses baseline points chosen outside the absorption lines, and sufficiently far from the lines centers. The stick spectrum (blue line of Fig. 6), calculated with the HITRAN database [15] agree well with the experiment both in intensities and wavelength positions.

6. Conclusion

The favorable situation described in section 5 with a slow relaxation laser does not seem to work for a fast relaxation laser as dye lasers. An experiment [16] made with a dye laser pumped by an Ar^+ laser modulated at a frequency $2\nu_{AO}$ close to the free spectral range ν_{RT} shows that: i) the sensitivity of ICLAS is low when the cavity length is tuned to the active mode-locked position ($2\nu_{AO}=\nu_{RT}$), ii) when the cavity length begins to detuned, the laser spectrum first condenses, then ICLAS is useless, iii) when the detuning further increases the spectrum becomes identical to that of the free running laser and the sensibility of the absorption is high again.

Our work demonstrates that the limitations of ICLAS with multimode lasers, due to their large coherence length, could be overcome with the use of a turbulent detuned actively mode-locked Ti:sapphire laser. The new technique TDAM-ICLAS has several advantages. The low coherence length of the TDAM laser eliminates harmful intracavity interferences and thus the indetermination of the spectral baseline which becomes stable and smooth. It is highly sensitive to intracavity absorption. We easily observed the very weak H_2O new line at 803 nm of [7]. We believe that the sensitivity is at least as good as that obtained with a free running multimode laser but without the problem of the spectral baseline. This is important for in situ measurements. We believe that the high sensitivity is due to the chaotic regime. It is well known that a chaotic system is very sensitive to internal disturbance. The precise dynamics of the TDAM laser should be subject of special study. In addition, our TDAM laser has no frequency chirp and therefore no absorption line broadening, and it is continuously tunable throughout the gain of the Ti:sapphire by just changing the inclination angle of the acousto-optical crystal.

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